

Article

Variable Angle Spectroscopic Ellipsometry Characterization of DMOAP-Functionalized Graphene Oxide Films

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Abstract: In our research, we investigated the optical properties of Graphene Oxide (GO) films functionalized with N,N-Dimethyl-N-octadecyl(3-aminopropyl)trimethoxysilyl chloride (DMOAP) using Variable Angle Spectroscopic Ellipsometry (VASE). We found that after the functionalization process, there was a notable increase in the thickness of the GO films. Additionally, there were observable changes in the Lorentz oscillator parameters, signaling modifications in the electronic transitions and structural properties of the films. Our results indicate that the functionalization with DMOAP not only impacts the electronic conjugation within the GO structures but also enhances the optical conductivity of the films, which is a finding of significant importance for potential applications in electronics, photonics, and materials science.

Keywords: graphene oxide; DMOAP; ellipsometry; functionalization

1. Introduction

Graphene oxide (GO) is a material known for its immense potential [1–6], largely attributable to its extensive surface area, cost-effective production, and capability for direct interaction with various biomolecules and nanoparticles [7]. GO is distinctively characterized by a combination of sp^2 and sp^3 hybridized carbon atoms [8]. Furthermore, its structure is enriched by various functional groups, including epoxy, carboxyl, and hydroxyl [9], making it an ideal substrate for functionalization processes.

GO finds applications in several fields. In biomedical applications, GO is particularly noted for its use in drug delivery systems [10]. This is due to its capacity to attach to and transport bioactive molecules, including specific anticancer drugs [11]. Research suggests that functionalized GO can deliver these drugs more efficiently than conventional methods. Moreover, its potential role in photothermal cancer therapies is currently under study [12]. In electronics, although GO is not inherently conductive, it is valued for its extensive surface area and compatibility with different solvents. This makes it a preferred choice for certain electronic components, particularly when solution-based methods are required [13]. In catalysis, GO’s inherent functional groups enable it to accelerate chemical reactions, benefiting sectors such as energy production and environmental applications [14]. In the field of composite materials, integrating GO into polymer matrices has been shown to improve the composite’s intrinsic properties, resulting in enhanced strength, flexibility, and electrical conductivity [15]. Although not the first choice material for energy storage, GO serves as a precursor to reduced Graphene oxide (rGO), which is used in the fabrication of high-performance supercapacitors [16,17].

A detailed study of GO’s optical properties, including its refractive index and extinction coefficient, is crucial for optimizing its utility across all mentioned applications and enhancing device performance in these fields.



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The optical properties of GO are greatly influenced by several key factors [18]. The intrinsic structural properties of GO, characterized by its mix of sp^2 and sp^3 hybridized carbon atoms and the presence of various functional groups, directly dictate its optical behavior. The degree of oxidation, which defines the types and amounts of oxygen-containing groups, has a direct impact on these properties. This influence is evident in the variations in absorption and reflectivity characteristics of GO, depending on its oxidation level. Additionally, the film thickness and uniformity of GO films are critical factors affecting their optical characteristics. Variations in thickness can lead to changes in light absorption and scattering, while non-uniformity can cause inconsistent optical behavior across the film. Furthermore, environmental factors, such as humidity, temperature, and ambient chemical exposure, can also induce reversible or irreversible changes in the optical characteristics of GO.

Among the methods for investigating the optical properties of materials, Variable Angle Spectroscopic Ellipsometry (VASE) offers several advantages [19,20]. It is a non-destructive technique that allows for precise measurements of optical constants and film thickness [20].

Schöche et al. [21] presented a detailed analysis of the optical constants of GO using spectroscopic ellipsometry. Their work distinctively employed a multi-location analysis approach, allowing for a precise determination of the anisotropic optical constants of GO layers. They also explored the intricate inter-band transitions in GO, linking these observations to theoretical models that consider varying levels of oxygen-containing functional groups on GO sheets.

Gangwar et al. [22] performed a thorough investigation of the optical behavior of GO when deposited on SiO_2/Si substrates. Their research explored key optical properties, such as bandgap, refractive index, extinction coefficient, and complex dielectric behavior, covering a broad wavelength range from 250 to 1650 nm.

Shen et al. [18] concentrated on characterizing the optical response of GO, particularly within the visible range, through spectroscopic ellipsometry. Their study was further enriched by incorporating the Lorentz oscillator model to analyze ellipsometric parameters in detail.

Our previous work [23] utilized the VASE technique to examine the optical response of GO in methanol films, which were dip-coated on glass substrates. This study provided valuable insights into the behavior of GO in different solvent environments, adding another dimension to our understanding of GO's optical properties. Moreover, in another previous article [24], we reported on a VASE characterization of GO thin films dip-coated on SiO_2/Si substrates and thermally reduced GO films in the 0.38–4.1 eV photon energy range.

The existing literature on the application of VASE to GO is indeed extensive, yet it has predominantly focused on the optical constants of GO and its reduced form [25]. However, the comparative analysis of GO in relation to its interaction with functionalization agents remains less explored.

A novel aspect of this research involves the functionalization of GO films with *n*-dimethyl-*n*-octadecyl-3-aminopropyltrimethoxysilyl chloride (DMOAP), a topic not previously explored, to the best of our knowledge. DMOAP molecules are known to form ordered self-assembled monolayers on oxide surfaces, with their long hydrophobic chains assuming an almost orthogonal orientation with respect to the substrate. These molecular monolayers have been extensively investigated for liquid crystal applications due to their ability to induce homeotropic alignment in nematic liquid crystals [26]. A relevant example is presented in [27], where a novel design for dye-doped liquid crystal (DDLC) microfluidic biosensing chips in polydimethylsiloxane material is revealed. In this study, DMOAP is used as an alignment layer in microfluidic channels, influencing the orientation of DDLCs and enabling the detection of antigen/antibody interactions with a sensitivity down to 0.5 $\mu\text{g/mL}$.

When DMOAP molecules are brought into contact with GO thin films, they interact with the oxygen-containing groups (hydroxyl, epoxy, and carboxyl) present on the GO surface. The silane end of DMOAP, susceptible to hydrolysis, forms silanols, which can subsequently condense with the hydroxyl groups on GO, forming stable covalent Si-O-C bonds. This process effectively grafts the DMOAP molecules onto the GO surface, modifying the surface chemistry and overall properties of the GO film.

Herein, we investigate the optical properties of DMOAP-functionalized GO films using VASE.

In the field of nanomaterials research, the functionalization of surfaces plays a pivotal role in tailoring their optical properties. Our study's focus on the use of DMOAP for modifying GO films finds a parallel in recent advancements in nanocluster science. Researchers [28] have employed (2,4-dimethylbenzenethiolate) DMBT to stabilize Ag₂₄Au nanoclusters, which are then post-synthetically modified with chiral ligands such as (R/S-1,1'-binaphthyl-2,2'-dithiol) R/S-BINAS to induce optical activity in initially achiral structures. This approach underscores a similar strategy in our work, where DMOAP serves not only as a functionalizing agent for GO but also as a means to manipulate its optical characteristics. Both methodologies, though applied to different materials and with distinct chemical agents, highlight the transformative impact of surface functionalization at the nanoscale.

While our study provides important insights, it represents only the initial steps in understanding the interactions between DMOAP and GO at a molecular level. Future research involving more in-depth experimental and theoretical approaches are needed to fully elucidate these interactions and their implications for material science, particularly in optoelectronics and liquid crystal display technologies.

2. Materials and Methods

In this study, SiO₂/Si substrates were first cleaned through a sequence of ultrasonic baths, using acetone, deionized water, and isopropanol, followed by subsequent air drying. Concurrently, an aqueous solution of GO (4 mg/mL) was prepared, involving 30 min of ultrasonic agitation to ensure uniform dispersion. This GO dispersion was then functionalized with DMOAP (Sigma Aldrich, St. Louis, MO, USA).

The functionalization occurred by adding DMOAP directly to the GO dispersion under continuous stirring. This step was essential for achieving a thorough and homogeneous mixing of DMOAP with the GO solution. The mixture was then left to react under ambient room temperature conditions, a procedure that did not require any additional catalysts, given the reactivity of DMOAP with the natural functional groups present on the GO surface, such as hydroxyl and carboxyl groups.

The deposition of GO films was carried out using a spin-coating technique, with the spin rate set at 1500 rpm. Thereafter, the optical properties of the films were examined using VASE.

Measurements of the ellipsometric angles, ψ and Δ , were conducted using a J.A. M2000 F (Woollam Co., Lincoln, NE, USA) rotating compensator ellipsometer (RCE), covering a wavelength spectrum from 300 nm to 800 nm. All measurements were carried out at room temperature at incidence angles of 60, 65, and 70 degrees.

The interpretation of the resulting data involved Lorentz optical models to determine the complex refractive index. In the data analysis stage, the Levenberg-Marquardt optimization algorithm was applied to minimize the Mean Square Error (MSE) within the VASE data.

We used WVASE32 software [29] to develop the optical models for our substrates.

This software employs advanced mathematical algorithms to provide rapid and precise data fitting. Beyond handling ellipsometric data, it also processes and simulates data related to reflectance and transmission. This includes the analysis of neutron reflectivity data, which is particularly useful for examining ultra-thin films, such as organic materials.

The MSE serves as a criterion for assessing the quality of the fit and is expressed as:

$$MSE = \sqrt{\frac{1}{2N - M} \sum_{i=1}^N \left[\left(\frac{(\psi_i^{mod} - \psi_i^{exp})}{\sigma_{\psi,i}^{exp}} \right)^2 + \left(\frac{(\Delta_i^{mod} - \Delta_i^{exp})}{\sigma_{\Delta,i}^{exp}} \right)^2 \right]} \quad (1)$$

Here, N denotes the total data points and M represents the fitting parameters' quantity. The terms $(\psi^{exp}, \Delta^{exp})$ and $(\psi^{mod}, \Delta^{mod})$ refer to the ellipsometric angles as observed and as predicted by the model, respectively, while σ_{ψ} and σ_{Δ} are the standard deviations of the observed ellipsometric angles. The predicted ellipsometric angles $(\psi^{mod}, \Delta^{mod})$ are determined by all the fitting parameters that constitute the multi-layered optical models.

3. Results and Discussions

Ellipsometric measurements for the uncoated silicon wafers were analyzed with a Si/SiO₂ model, applying the silicon and silica optical constants available in the instrument's software, namely Si.jaw and SiO₂.jaw. This model revealed that the native oxide layer on the silicon was approximately 2 nm thick.

DMOAP-functionalized GO films were analyzed using a sum of three Lorentzian oscillators. The complex dielectric function can be expressed as:

$$\tilde{\epsilon}(h\nu) = \epsilon_1 + i\epsilon_2 = \epsilon_{\infty} + \sum_{k=1}^N \frac{A_k}{E_k^2 - E^2 - i\Gamma_k E} \quad (2)$$

In this equation, E represents the photon energy of incoming light, ϵ_{∞} denotes the real part of the dielectric function as E approaches infinity, A_k is the oscillator strength, Γ_k represents the broadening constant, and E_k is the resonant energy of the k -th oscillator. The term A_k also quantifies the contribution of each k -th oscillator to the overall system.

Parameters such as film thickness and the Lorentzian oscillator parameters were set as variables to be adjusted during the fitting process.

Figure 1a displays the experimental and simulated values for the ellipsometric parameter ψ , while Figure 1b shows those for Δ in DMOAP-functionalized GO films.

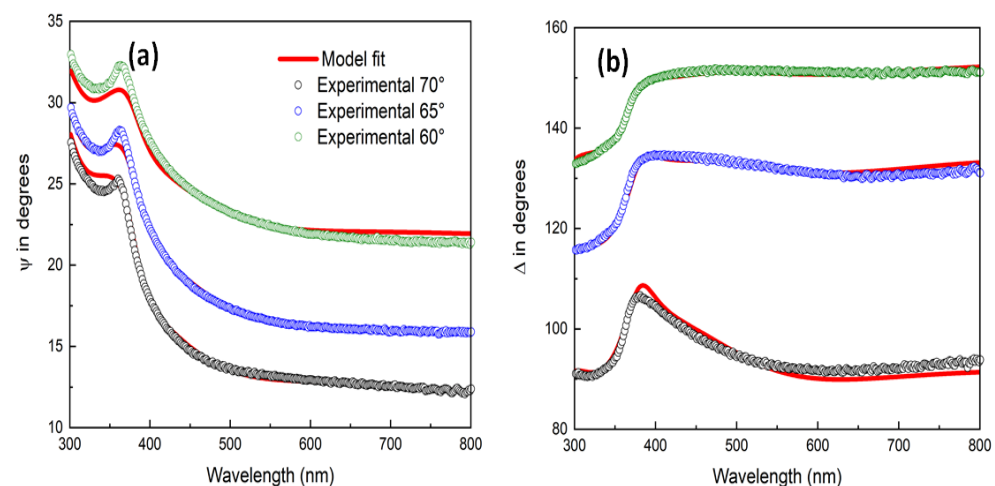


Figure 1. Variable angle spectroscopic ellipsometry of DMOAP-functionalized graphene oxide films on SiO₂/Si substrates. Comparison of experimental and simulated (a) ψ and (b) Δ at 60°, 65° and 70° incident angles.

The data generated by our model showed a high degree of concordance with the values we acquired experimentally, as can be seen in Figure 1.

Table 1 shows the best fit parameters for DMOAP-functionalized GO films on SiO₂/Si, with a low MSE around 7. The table also includes Lorentz oscillator parameters for GO films from earlier research [24] for comparison.

Table 1. Parameters of Lorentz oscillators for DMOAP-functionalized graphene oxide films (current study) compared to graphene oxide films (referenced in [24]). The unit for Amplitude A_k is eV², while both the central energy E_k and broadening Γ_k are measured in eV. The film thickness ‘d’ is expressed in nm, and the high-frequency dielectric constant ϵ_∞ is without dimension.

	DMOAP-Functionalized GO	GO [24]
d (nm)	46 ± 1	19 ± 1
ϵ_∞	1.0 ± 0.1	1.62 ± 0.04
A_1 (eV ²)	8.1 ± 0.4	1.8 ± 0.2
Γ_1 (eV)	0.39 ± 0.02	1.05 ± 0.09
E_1 (eV)	3.3 ± 0.1	2.8 ± 0.1
A_2 (eV ²)	7.4 ± 0.7	7.0 ± 0.2
Γ_2 (eV)	1.0 ± 0.1	0.69 ± 0.05
E_2 (eV)	4.1 ± 0.1	3.22 ± 0.01
A_3 (eV ²)	13.0 ± 0.5	2.3 ± 0.1
Γ_3 (eV)	2.1 ± 0.2	0.59 ± 0.08
E_3 (eV)	3.1 ± 0.1	3.90 ± 0.02

The functionalization of GO with DMOAP significantly alters its physical and electronic properties, as evidenced by the changes in the Lorentz oscillator parameters. When comparing DMOAP-functionalized GO with its pristine counterpart, several notable differences are observed. The thickness of the DMOAP-functionalized GO film increases significantly, more than doubling compared to the pristine GO film. The DMOAP-functionalized GO films show a 142% increase in thickness relative to the non-functionalized GO films, indicating the successful addition of DMOAP layers. This increase is attributed to the DMOAP molecules’ covalent bonding, which contributes to the overall structural dimension of the film. Equally important is the observation that the high-frequency dielectric constant ϵ_∞ for the DMOAP-functionalized GO exhibits a reduction compared to pristine GO. The altered electronic environment, resulting from the introduction of DMOAP, appears to influence how the material responds to electric fields at high frequencies, indicating modified dielectric properties. Regarding the Lorentz oscillator parameters—amplitude, broadening, and center energy—the DMOAP-functionalized GO presents higher amplitudes, suggesting enhanced electronic transitions within the material. The broadening parameter, indicative of the range of energy states or the level of disorder, is wider in the functionalized GO. This broadening suggests that the DMOAP functionalization introduces a degree of structural and electronic heterogeneity, likely arising from the diverse sites at which the DMOAP molecules attach. While the center energies are relatively consistent between the DMOAP-functionalized and pristine GO, slight shifts can be seen. These shifts are indicative of the subtle changes in the electronic structure after functionalization, affecting the resonant frequencies of the oscillators.

The functionalization of GO with DMOAP is characterized by a complex interplay of forces and bonding mechanisms.

In this study of GO film functionalization with DMOAP, we draw parallels from the principles outlined in the research by Tessonier et al. [30] on the dispersion of alkyl-chain-functionalized rGO sheets in nonpolar solvents. Their work demonstrates how the grafting of alkyl chains onto rGO significantly enhances its dispersion in nonpolar solvents, due to the strong interactions between the alkyl chains and the carbon lattice of rGO. Applying similar concepts to our study, it is hypothesized that the large aromatic

system in GO might facilitate π - π stacking interactions with the hydrophobic alkyl chains of DMOAP. Additionally, van der Waals forces could also play a significant role in the interaction between the hydrophobic tails of DMOAP and the basal plane of GO. The hydrophilic regions of GO, owing to the oxygen-containing functional groups, can interact with the hydrolyzed silane head of DMOAP through hydrogen bonding or even covalent bond formation. This occurs as the silanol groups (formed after the hydrolysis of DMOAP) undergo condensation reactions with the oxygen-containing groups on GO. This interaction may lead to a redistribution of electron density, affecting the delocalization of π -electrons and thus the electronic conjugation. In addition, functionalizing GO with DMOAP could also introduce new energy levels or modify existing ones in the electronic structure of GO. This happens as the molecular orbitals from DMOAP interact with the carbon lattice of GO. However, the precise nature of GO-DMOAP interactions at the molecular level remains to be fully explored. Future experimental and theoretical studies will be necessary to validate and expand upon these initial observations.

Figure 2 illustrates the estimated dispersion relations, as determined through VASE measurements for DMOAP-functionalized GO films.

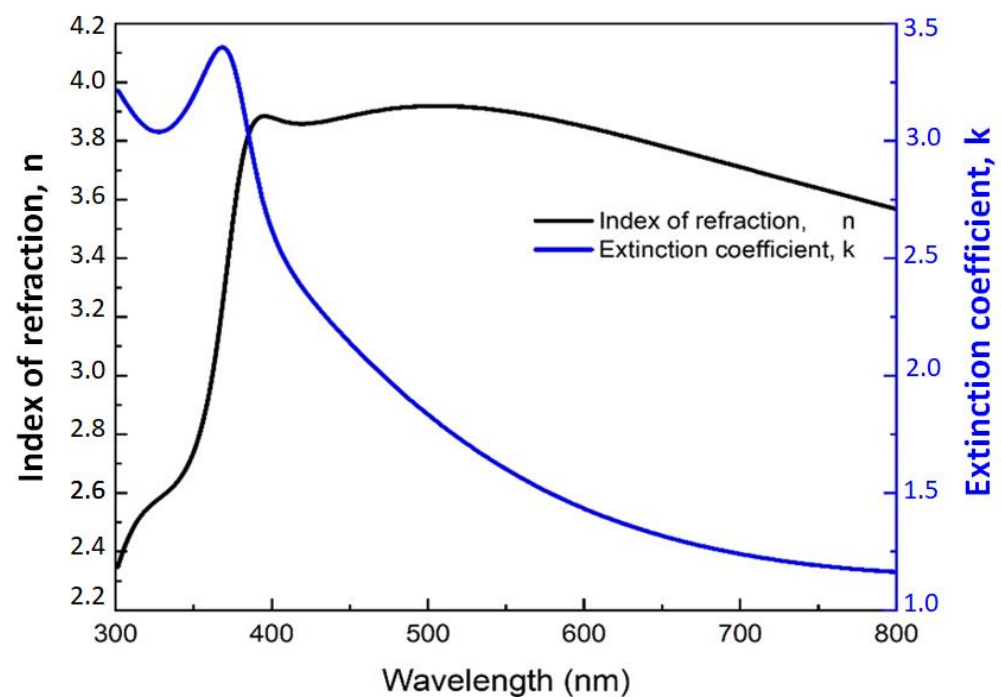


Figure 2. Estimated dispersion laws of the DMOAP-functionalized graphene oxide films. The black curve illustrates the index of refraction (n), while the blue curve represents the extinction coefficient (k).

The complex optical conductivity $\sigma = \sigma_1 + \sigma_2$ is related to the complex dielectric constant $\varepsilon = \varepsilon_1 + \varepsilon_2$ by the following relations [31]:

$$\sigma_1 = \omega \varepsilon_2 \varepsilon_0 \text{ and } \sigma_2 = \omega \varepsilon_1 \varepsilon_0 \quad (3)$$

where ω is the angular frequency, and ε_0 is the free space dielectric constant.

As seen in Figure 3, DMOAP functionalization increases the optical conductivity of GO. This enhancement is clearly illustrated when comparing the untreated GO films (Figure 3a) with those treated with DMOAP (Figure 3b).

GO has disrupted sp^2 hybridization due to the presence of various oxygen-containing functional groups. When DMOAP is introduced, it reacts with these groups, leading to a partial restoration of the electronic conjugation system typical of pristine graphene. This restoration occurs through the removal or reduction of oxygen-containing groups, thereby

increasing the optical conductivity. Moreover, DMOAP molecules have long alkyl chains and a functional head that can interact with the surface of GO. This interaction modifies the electronic environment at the surface of the GO, potentially reducing the scattering of charge carriers and stabilizing the material. Enhanced charge carrier mobility typically leads to an increase in optical conductivity.

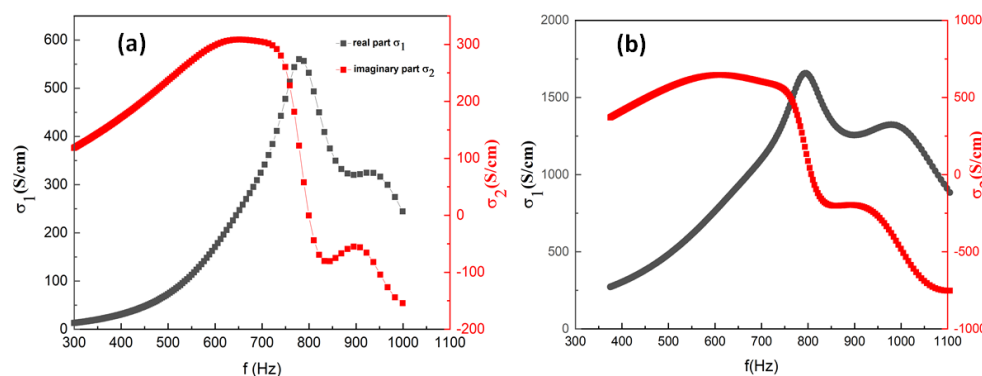


Figure 3. Optical conductivity variation with frequency for both (a) graphene oxide films and (b) graphene oxide films treated with DMOAP on SiO₂/Si substrates.

The variation in optical conductivity is particularly significant because it directly impacts the potential applications of these materials in optoelectronic devices. The increased optical conductivity in DMOAP-functionalized GO films suggests enhanced electron mobility, which is a desirable characteristic in applications such as photodetectors, solar cells, and other electronic components.

4. Conclusions

Our research highlights the significant impact of DMOAP functionalization on the optical properties of GO films using VASE, a subject previously not explored in the scientific literature.

One of the key discoveries of this research is the notable increase in the thickness of GO films post-functionalization with DMOAP. This increase is more than double compared to pristine GO films, indicating the successful integration of DMOAP through covalent bonding. This bonding significantly contributes to the overall structural dimensions of the film, thereby profoundly altering both its physical and electronic properties.

The study also observes distinct changes in the high-frequency dielectric constant and the Lorentz oscillator parameters of the DMOAP-functionalized GO films. These changes suggest markedly enhanced electronic transitions within the material and indicate a substantially modified dielectric property. The broadening parameter in the functionalized GO, indicative of the range of energy states or the level of disorder, is also found to be wider, implying that DMOAP functionalization introduces a substantial degree of structural and electronic heterogeneity.

Moreover, this research draws parallels from other scientific studies to hypothesize the complex interactions at play in the functionalization process. It is proposed that the large aromatic system in GO might facilitate strong π - π stacking interactions with the hydrophobic alkyl chains of DMOAP. Additionally, van der Waals forces and potential hydrogen bonding or covalent bond formation between the silanol groups (formed after hydrolysis of DMOAP) and the oxygen-containing groups on GO are deemed crucial in this interaction.

In addition to these physical interactions, the study suggests that the molecular orbitals of DMOAP may interact with GO's carbon lattice, leading to a significant reconfiguration of the electronic structure. This interaction could introduce new energy levels or modify existing ones in GO, profoundly affecting the delocalization of π -electrons and thereby impacting the electronic conjugation within the material.

Furthermore, our study provides an in-depth analysis of the dispersion relations and optical conductivity of DMOAP-functionalized GO films. It is observed that the functionalization markedly increases the optical conductivity of GO, which is a critical factor for its potential applications in optoelectronic devices. The increased optical conductivity strongly hints at an enhanced electron mobility, a desirable characteristic in applications such as photodetectors, solar cells, and other electronic components.

While our results suggest changes in the electronic structure and bonding, these are preliminary observations. To fully elucidate the molecular interactions, further in-depth experimental and theoretical studies are essential to validate the hypothesized interactions between DMOAP and GO. Moreover, our research proposes that the GO functionalization with DMOAP may confer improved stability against environmental factors such as humidity and temperature, which are known to affect the integrity of GO. However, a comprehensive evaluation, including long-term stability tests under various conditions, is essential to confirm these benefits. Additionally, future research works should investigate the environmental implications of these functionalized films, including aspects such as biodegradability and potential ecotoxicity.

In conclusion, our work paves the way for customizing the properties of DMOAP-functionalized GO films. The substantial enhancement in optical conductivity observed in these films opens up new frontiers in various technological applications, particularly in optoelectronics and photonics. In the field of optoelectronics, DMOAP-functionalized GO films could play a vital role in developing high-efficiency photodetectors and advanced light-emitting diodes. The enhanced optical conductivity also promises significant advancements in energy technologies, such as solar cells, where it could lead to increased efficiency in photovoltaic devices. Furthermore, the improved optical properties make these materials ideal candidates for transparent conductive coatings, which are essential in next-generation display technologies and touchscreens. In the field of photonics, these functionalized films could be crucial in developing waveguides and optical sensors, offering an enhanced performance due to their improved interaction with light. Additionally, the application of these films in liquid crystal devices holds particular promise. Their improved optical conductivity and altered surface properties could lead to a better control and alignment of liquid crystals, crucial for the performance and efficiency of liquid crystal displays and related technologies.

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Abbreviations

GO	Graphene Oxide
DMOAP	N,N-Dimethyl-N-octadecyl(3-aminopropyl)trimethoxysilyl chloride
VASE	Variable Angle Spectroscopic Ellipsometry
rGO	Reduced Graphene Oxide
DDLC	Dye-Doped Liquid Crystal
MSE	Mean Square Error
RCE	Rotating Compensator Ellipsometer

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